Crystallization of pentagon packings

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Abstract. We present an experiment of densification of a 2D horizontal assembly made of regular pentagons. The annealing leads to a dense crystalline arrangement with quasisixfold symmetry in spite of the geometrical characteristics of the grains and of the *a priori* short-range correlation length in the packing. This configuration yields the maximum average number of side to side contacts between the grains and probably the maximum packing fraction.

1. Introduction

Two-dimensional packings of discs have been used to model a large number of systems ranging from (two-dimensional) dense gases to solids, with liquids and 'granular media' as intermediate steps. The structure of 2D dense packings has been intensively studied by numerical simulations (Visscher and Bolsterli 1972), experimentally (Dodds 1975, Bideau *et al* 1986) and theoretically (Rubinstein and Nelson 1982). The experimental study of Quickenden and Tan (1974) and that of Lemaître *et al* (1991) on packings on a blowing air table have shown the structural changes when densifying packings of discs. The particle size distribution has been used in order to understand the short-range geometrical correlations (Troadec *et al* 1994).

Systems with more complicated particle geometry were also considered. Some numerical (Cuesta and Frenkel 1990) and theoretical results (Vieillard-Baron 1972) are available on ellipses. Ammi *et al* (1987) have studied the geometry and the mechanical properties of packings of various regular polygons, built under pseudocentral force. Polygons are particular because they present two types of contact: side to side contacts and side to vertex contacts.

The blowing air table used by Lemaître *et al* (1991) for discs allows also the study of the influence of the shape of particles on the structure of the packing. The first tests were performed with ellipses, but, as the correlation length in the ordered zones was always larger than the size of the system, no definitive conclusions could be obtained. Following the work of Ammi *et al* (1987), and looking for particles which should not lead to any long-range order, we have chosen pentagons.

We present, in this paper, a study of the densification of a packing of pentagons on the blowing air table. Surprisingly, the simulation of the annealing of the packing leads to a crystal. In the following, we describe the experiment. The structure of the theoretical crystal is then described, and compared to the experimental one.

2. Experimental conditions

The experimental device has already been described in detail (Lemaître *et al* 1990). A wind tunnel blows a uniform vertical air stream through a horizontal porous square plate (useful surface 2000 cm^2) made of sintered bronze. The air stream, controlled by the voltage applied to two air fans, sustains small light particles. Thus it is possible to obtain 2D assemblies in which the friction with the table does not prevent local arrangements. Because of the local fluctuation of the air velocity field, the particles rearrange permanently and after a short 'thermalization' time, their spatial distribution becomes homogeneous. The particles are submitted to hard-core interactions, but also to weak hydrodynamic interactions that cause an effective short-range order repulsion (Lemaître *et al* 1991).

In our experiments the assemblies are made of 2200 styrene pentagons of 6 mm sidelength and 1 mm thickness. The horizontality of the table has been adjusted for a voltage $V = 150$ V applied to the air fans. With such a voltage, the pentagons are distributed over the whole surface of the table and form a not very dense assembly (packing fraction $C \sim 0.7$). The simulation of annealing is obtained by decreasing the voltage V from 150 V, very slowly in order that the system remains in equilibrium with the air flow. As the voltage decreases, the table bends down because of the diminution of the air pressure under it (the table deformation is about 10 μm for a variation of 200 V of V). There is also probably a modification of the hydrodynamic interactions. The particles tend to gather towards the centre of the table. Thus, the structure of the assembly evolves and its packing fraction, the ratio of the area of the pentagons to the area they occupy effectively, increases.

In order to study that evolution, video records and snapshots are taken. Both of them can be digitized and studied with an image processing system on a workstation.

3. Experimental results

Figures 1 and 2 show respectively the variations of the packing fraction and the variations of the velocity of the particles when one decreases the voltage V. At the beginning of the experiment, when a strong air stream is applied, the particles move quickly. Their motion occurs on all degrees of freedom ; in particular their rotation speed is high. There is no orientational correlation between the particles. If we consider the particle positions, no appreciable difference between discs and pentagons can be observed in this regime. The Fourier transform of particle positions performed at $V = 100$ V (figure 3(a)) shows that the assembly is isotropic and disordered.

When we reduce slowly the air stream, the packing fraction increases, but due to the hydrodynamic short-range repulsion, there is no real contact between the particles. In one sense, this repulsion smooths the geometry of the particles. The environment of one particle remains close to that of a disc in a disc packing. Below approximately $V = 90$ V, ordered zones with sixfold symmetry appear, separated by dislocation lines or amorphous zones in permanent motion. At that stage, order affects only the particle positions. Figure 3(b) gives the Fourier transform of the position of the centres for $V = 60$ V.

When the air stream is further reduced, an orientational order appears which leads to a distortion of the lattice, and we obtain crystalline zones (figure 4). One single crystal cannot be obtained because the basic cell is not quite isotropic. The crystalline zones grow by nucleation within the previous ordered zones. It takes a few minutes to get a crystalline zone big enough to be studied. Note that two mechanisms are involved in the formation of the structure:
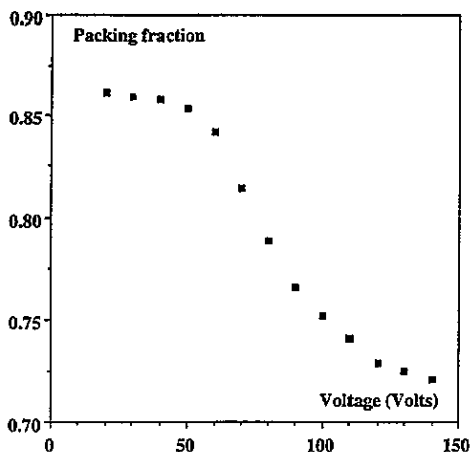
Figure 1. Variation of the packing fraction C with the voltage V applied to the air fans for a packing of 2200 pentagons. At low voltage, the packing fraction does not reach the value 0.92 that we will find in crystalline zones because it is measured in the centre of the table where the packing defects are more numerous (see figure 4).
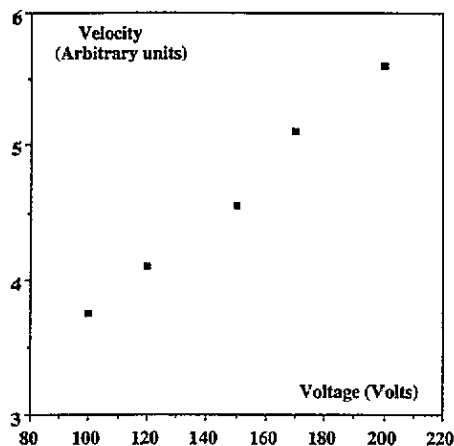
Figure 2. Variation of the mean velocity of particles with the voltage V applied to the air fans.

(i) a decrease of the velocity of the particles (figure 2), which is equivalent to a decrease of the 'granular' temperature of the packing,

(ii) an increase of the packing fraction at the centre of the table (figure 1).

Both are necessary to get a crystal. No crystal is obtained at both high packing fraction and high air stream.

An experiment close to ours was performed some years ago (Spaepen and Nelson unpublished, quoted by Sachdev and Nelson 1985), in which the density of pentagons was gradually increased in a vibrating shaking table apparatus. Although a domain with the same crystalline structure as ours could be observed, the emphasis was put only on the fact that the centres of the pentagons formed a hexagonal lattice.

Qualitatively, the crystalline structure of our experiment is favoured by two antagonistic effects, which both increase when the 'thermal' motion is reduced—the compaction of the packing and the hydrodynamic repulsion between the particles: there is no real contact between the polygons. The repulsion is stronger when the number of side to side neighbourhoods is maximum. If one discards the possibility of vertex to vertex neighbourhood which should be unstable in a dynamical system, it can be shown (see appendix) that, in a dense packing, the largest value of the mean number of side to side neighbourhoods of a pentagon is four. In the crystalline zones, this is precisely the value observed for each pentagon which has also two side to vertex neighbourhoods.

Generally, even after a long time, the ordered zones (with quasi sixfold symmetry) are not perfectly crystalline. In some places there is no orientational order. In other places there are more localized orientational defects. As we can see in figure 5, in a crystalline zone, there is a particular direction a along which lines of pentagons with side to vertex neighbourhood are directed. The orientation of a pentagon can be identified by the angle a between the direction a and a line pointing from the centre of the pentagon to the vertex
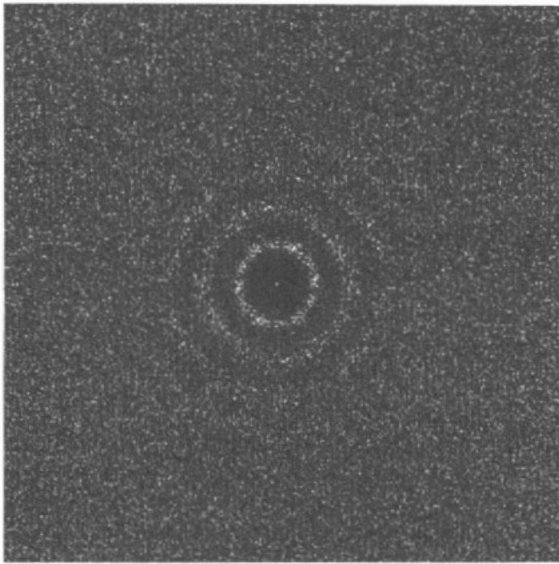
(a)
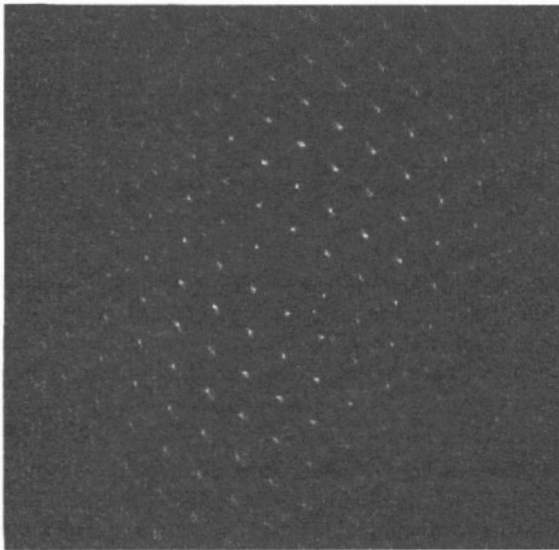
(b)

Figure 3. Fourier transform of the positions of the centres of the pentagons (a) at $V = 100$ V, (b) at $V = 60$ V.

closest to direction a. This has meaning only where there is a nearly perfect orientational order. While considering orientation of pentagons, we have to keep in mind that even at a low voltage (less than 40 V) pentagons still oscillate around their equilibrium direction. When we take a snapshot of the packing, we freeze positions at a given time. Some of the observed misorientation of particles can be explained by the fact that we do not average orientations over the time. So, we consider that there is no orientational defect when the
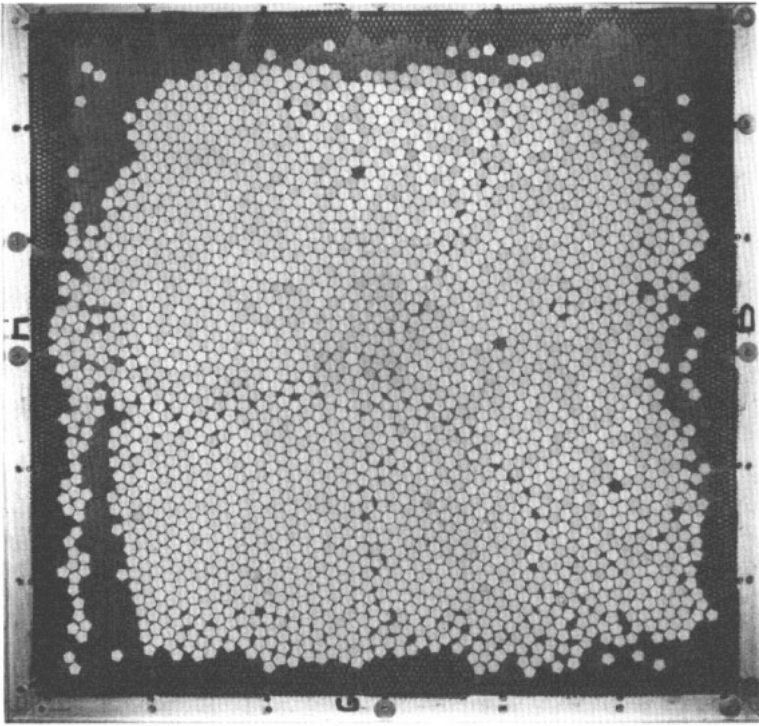
Figure 4. An experimental crystal of pentagons on the blowing air table.

angle a of orientation of a pentagon is less than $\pi/10$.

All crystalline areas stop either on the limit between two ordered zones with quasisixfold symmetry, or, within the same ordered zone, on a line of orientational defects or a zone without orientation order. On a line of defects, there is a switching of crystal orientation. Orientation switchings are of 180° or of approximately 60°. Boundaries of oriented areas are well defined. There is no isolated orientational defects. As one misoriented pentagon is separated from a stable position by less than $\pi/10$ radians and as there is no static friction, nothing stabilizes the pentagon outside a crystallographic position. Thus there is no metastable position between two orientations separated by $\pi/5$. As there is no static friction, all defects have to be stabilized by their environment.

4. Description of the crystallized structure

From the structure observed in the experimental packing, we have drawn the regular lattice of figure 6. That structure (among some others) has already been briefly quoted by Henley (1986) who thinks of it as the closest packing of pentagons. As said above, there is a principal direction a in the packing, along which pentagons are pointing. The two other directions b and c are determined from the side to side contacts. The cell parameters of the theoretical lattice are $a = 1.538\,84s$, $b = 1.410\,64s$, $c = 1.464\,39s$ where s is the sidelength of the pentagons. There is some distortion between these parameters: $(a - b)/b = 9.1\%$ between a and b, $(a - c)/c = 5.1\%$ between a and c and $(c - b)/b = 3.8\%$ between b and c. The angles are slightly different from 60°: more
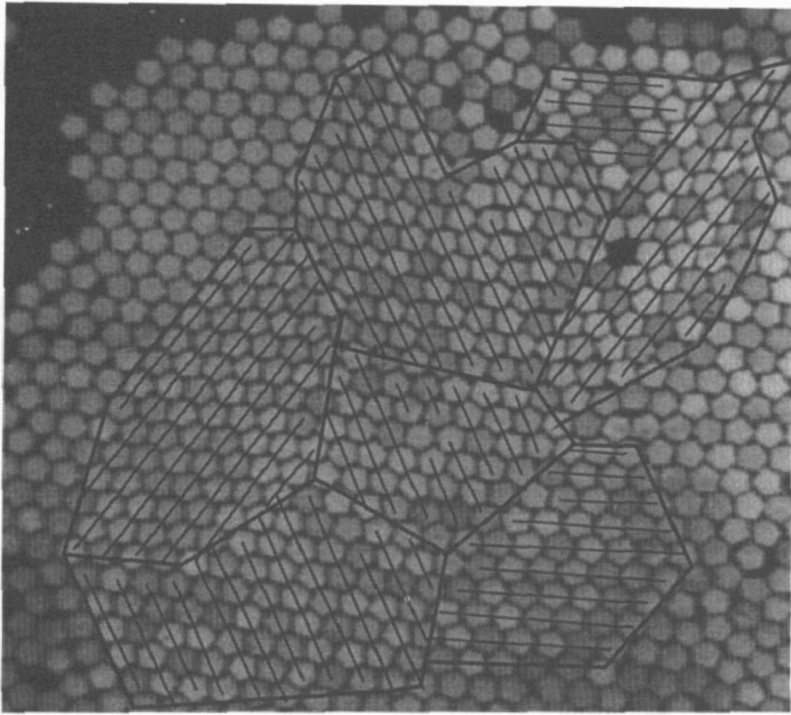
Figure 5. Visualization of the limits of the crystalline zones in the top left part of the packing of figure 4. The thin lines give the direction a in the different zones.

precisely, $(a, b) = 59.3°$, $(a, c) = 56.0°$, $(b, c) = 64.7°$. So, the difference between disc and pentagon leads to a reduction of the symmetry from sixfold to rectangular, and the 2D space group of the arrangement in figure 6 is *pm* (*International Tables for Crystallography* vol A). Moreover, the space group is almost *p2mg*, a subgroup of the 2D hexagonal space group *p6mm*, but the glide line *g* and the twofold rotation point are not true symmetry elements. The packing fraction of the regular packing of figure 6 is $C = 0.9213$, while that of the structure with *p2mg* symmetry is only 0.8389. Actually, because of the hydrodynamic repulsion between the particles, the experimental structure is intermediate (the experimental values of angles (a, b), (a, c) and (b, c) are closer to 60° than the values given above).

In the packing of figure 6, each pentagon is surrounded by $z = 6$ pentagons. There are $z_{ss} = 4$ side to side contacts and $z_{sv} = 2$ side to vertex contacts. We prove in the appendix that—provided no vertex to vertex contact exists—the values six and four are the largest values that can be obtained respectively for the average coordination number, z, and the average number, z_{ss}, of side to side contacts per pentagon in any assembly (regular or disordered) made of regular pentagons:

$$z \leqslant 6 \tag{1a}$$

$$z_{ss} \leqslant 4. \tag{1b}$$

The first inequality is actually more general and holds even when vertex to vertex contacts take place or for higher-order polygons (heptagons, ...). Moreover, it may be proved that this average maximum is also a *local* maximum (which is not true for z_{ss}). Coming back to our pentagons, it may be proved conversely that when both maxima are reached
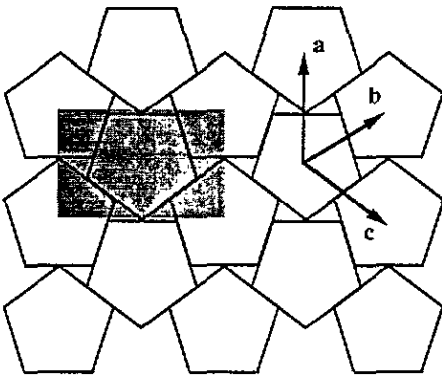
Figure 6. The theoretical crystal of pentagons. The space group is *pm*. The unit cell is shown in grey.

simultaneously, the pore space consists of triangles only, each with two side to side contacts. When the length of one side of a triangle is fixed, the minimal area of the triangle is obtained, with the constraint of angles $\leqslant 2\pi/5$, when two angles are equal to $2\pi/5$ and the third is equal to $\pi/5$. Minimization of the area is still better when the side to vertex contact is in the middle of the side. Then, our crystal has the largest possible packing fraction with $z = 6$ and $z_{ss} = 4$. We did not succeed in proving that this is the largest possible value.

5. Conclusion

Starting from an isotropic assembly of regular pentagons on an air table, we obtain by progressive densification and cooling a crystalline arrangement with quasisixfold symmetry in spite of the shape of the grain. The three directions are slightly distorted. Defects arise along lines.

This configuration maximizes the average number of contacts and the average number of side to side contacts. This may be proved theoretically provided no vertex to vertex contact exists, which is the case on the air table. The packing fraction is probably maximum too, but we were just able to get partial results.

The importance of side to side contacts at large packing fraction is due to the possibility for permanent reorganization on the table. It would not be the case in the presence of friction (Ammi *et al* 1987). We are presently studying a numerical model using molecular dynamics. Pentagons are placed randomly with random orientation inside a large circle. Then a densification towards the centre of the circle is performed. After relaxing the packing, one gets crystallized zones.

Appendix

We consider a packing of regular pentagons dense enough so that each pentagon has at least two contacts. We do not require the packing to be crystalline but we discard the possibility of vertex to vertex contacts. This packing may be regarded as a graph made of polygons:

(i) the P pentagons which form the grain space of the packing;

(ii) T triangles and P_n polygons with n sides (n-gons, $n \geqslant 4$), which form the pore space. The n-gons are concave as some vertices belong neither to a side to side nor to a side to vertex contact. Let $\phi(n)$ be the mean number of 'concave' vertices of an n-gon. The corresponding angles of the n-gon are $7\pi/5$. Of course, we have:

$$0 \leqslant \phi(n) \leqslant n - 3 \qquad (A1)$$

(ii) moreover, the D_2 side to side contacts between the pentagons will be represented as 2-gons (figure A1).
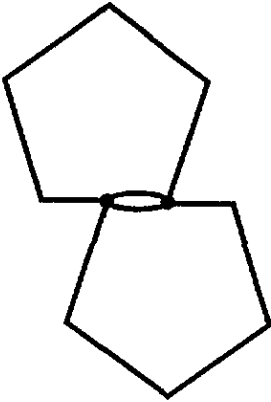

Figure A1. The side to side contacts between the pentagons can be represented as 2-gons with curved sides.

A1. Topological properties: proof of inequality (1a)

Some general relations exist between the elements of the graph. The total number N of polygons is

$$N = P + T + D_2 + \sum_{n \geqslant 4} P_n. \qquad (A2a)$$

The number of vertices is

$$V = 5P. \qquad (A2b)$$

The coordination number of the 'concave' vertices of an n-gon is two, while that of all the other vertices is four. The number E of edges of the graph is then

$$E = 2V - \sum_{n \geqslant 4} \phi(n) P_n. \qquad (A2c)$$

We are mainly interested in quantities related to the pentagons of the packing. According to the type of contact, the mean coordination number z of the pentagons can be written as

$$z = z_{ss} + z_{sv}$$

where z_{ss} and z_{sv} are the mean numbers of respectively side to side and side to vertex contacts around a pentagon. We have some relations among the different polygons of the graph that can be given as a function of P, z_{ss} and z_{sv} only:

$$D_2 = z_{ss} P/2. \qquad (A3a)$$

Euler's law gives

$$T + \sum_{n \geqslant 4} P_n = \left(\frac{z_{ss} + z_{sv}}{2} - 1 \right) P. \tag{A3b}$$

Counting the polygons forming the pore space of the packing, we can express V as:

$$V = 3T + \sum_{n \geqslant 4} n P_n - \frac{z_{sv}}{2} P.$$

Then, from (A2b) and (A3b):

$$\sum_{n \geqslant 4} (n - 3) P_n = \left(8 - \frac{3z_{ss} + 2z_{sv}}{2} \right) P. \tag{A3c}$$

The number of edges of the graph can be expressed as

$$E = (5 + z_{ss} + z_{sv}/2) P.$$

Then, from (A2b) and (A2c):

$$\sum_{n \geqslant 4} \phi(n) P_n = \left(5 - z_{ss} - \frac{z_{sv}}{2} \right) P. \tag{A3d}$$

Actually, the same derivation may be performed when introducing vertex to vertex contacts or when replacing pentagons of the packing by higher-order polygons. Moreover, because of the right inequality in (A1), we get (1a) from (A3c) and (A3d):

$$z = z_{ss} + z_{sv} \leqslant 6.$$

A2. Metric properties: proof of inequality (1b)

We can now use the peculiar values of the angles in regular polygons to refine inequality (A1), at least when no vertex to vertex contact exists. In an n-gon, concave angles are $7\pi/5$ and side to vertex angles are less than $2\pi/5$. The sum of the angles is

$$(n - 2)\pi = \frac{7\pi}{5} \phi(n) + \frac{2\pi}{5} \alpha(n) + \sum_i \beta_i(n) \tag{A4}$$

where $\alpha(n)$ is the number of side to side contacts, $\sum_i \beta_i(n)$ is the sum of the angles less than $2\pi/5$ (their number is $n - \phi(n) - \alpha(n)$). The possible values of $\phi(n)$ are $\phi(4) = 1$, $\phi(5) = 1$ when all acute angles are equal to $2\pi/5$ or $\phi(5) = 2$, $\phi(6) = 2$, $\phi(7) = 3$, $\phi(8) = 3$ or $4 \dots$.

We first note that from (A4)

$$\phi(n) \leqslant \tfrac{5}{7}(n - 2). \tag{A5}$$

Setting now $\beta_i(n)$ to its maximum value $2\pi/5$, we get another inequality

$$\phi(n) \geqslant \frac{3n}{5} - 2.$$

Note that $(n - 3)/2 \leqslant 3n/5 - 2$ as soon as $n \geqslant 5$. More generally, as $\phi(n)$ is an integer for a given n-gon, we have the important result which improves (A1):

$$\frac{n - 3}{2} \leqslant \phi(n) \leqslant n - 3$$

when $n \geqslant 4$.

Now, because of the left inequality, we get inequality (1b) using (A3c) and (A3d):

$$z_{ss} \leqslant 4.$$

A3. Application to our crystal

Our packing is characterized by $z = 6$ and $z_{ss} = 4$ which, according to inequalities (1a) and (1b) are the maximum possible values.

From (A3c) and (A3d), the equality $z = 6$ implies

$$\sum_{n \geqslant 4} (n - 3) P_n = \sum_{n \geqslant 4} \phi(n) P_n$$

which means that either $\phi(n) = n - 3$ or $P_n = 0$. Strictly speaking, the equalities hold 'almost everywhere', i.e. the exceptions are negligible compared to the mean behaviour in the graph; this is the case for a crystal. From (A5), since $\frac{5}{7}(n - 2) < n - 3$ as soon as $n \geqslant 6$ we deduce that $\phi(n)$ can be equal to $n - 3$ only when $n = 4$ or 5 (and two concave angles in the pentagons). Then, for $z = 6$, we have

$$P_4 + 2P_5 = \frac{4 - z_{ss}}{2} P \qquad T + P_4 + P_5 = 2P.$$

From (A3c) and (A3d), the equality $z_{ss} = 4$ implies that the pore space consists of triangles and concave pentagons only with all acute angles equal to $2\pi/5$, with

$$T + P_5 = \left(1 + \frac{z_{sv}}{2}\right) P$$

$$P_5 = \left(1 - \frac{z_{sv}}{2}\right) P \qquad T = z_{sv} P.$$

Then, for $z = 6$ and $z_{ss} = 4$, we have only triangles in the pore space, which is effectively realized in our packing.

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